Phase Equilibria Liquid-Liquid and Liquid-Vapor in the Binary n-Hexane – Water System[1]

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1 Paper presented at the Fourteenth Symposium on Thermophysical Properties, June 25-30, 2000, Boulder, Colorado, U.S.A.

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ABSTRACT

Experimental investigations of the $PVTx$ – properties in the n-hexane – water system with the different compositions (x) 0.166, 0.257, 0.347, 0.615, 0.827, 0.935, and 0.964 molar fraction (m.f.) H_2O were made with a piezometer of constant volume. Measurements have been carried out along isochores at densities 66-834 kg·m^{-3}, temperatures 372-620 K, and pressures up to 65 MPa. The phase diagrams liquid-liquid and liquid-vapor are constructed.

For the description of the thermodynamic properties of the n-hexane – water binary system, the equation of state of Soave-Redlich-Kwong and scaling equations are used.

KEY WORDS: binary mixture; critical exponent; equation of state; liquid-liquid; liquid-vapor; n-hexane; power law; PVT; water.

1. INTRODUCTION

The research of thermodynamic properties of the binary n-hexane – water system has both a practical, and a wide theoretical interest in sense of analysis of a system behavior not blending in usual condition. The PVT-properties of this system were studied by a number of the authors, but all researches fall into areas with high concentration of water and pressures above 20 MPa [1-5]. In this work the results of experimental investigations of P, V, T, x (pressure, volume, temperature, composition) properties are reported for the binary n-hexane – water system. The measurements have been carried out in the range of temperatures 372-620 K, densities 66-834 kg/m^3 and at pressures up to 65 MPa for 7 values of composition: 0.166, 0.257, 0.347, 0.615, 0.827, 0.935, and 0.964 molar fraction (m.f.) H_2O.

The measurements encompass the area of thermodynamic parameters between phase equilibrium liquid-liquid and liquid-vapor.

The studies of PVTx – properties were made on special apparatus, by the piezometer constant volume method. . For each composition the measurements were carried out along 7-9 isochores in a wide range of densities.

2. EXPERIMENTAL

The main part of the apparatus is a cylindrical vessel made of corrosion-proof steel (12X18H10T) with internal diameter 8.721 mm. The temperature was regulated automatically with the help of thermoregulator VRT-3 with an accuracy of 0.01 K. The pressure was measured with a weighted-piston manometer of accuracy 0.05% with the help of a membrane zero-sensor. The volume of the piezometer was determined on water and is peer to 21.160±0.025 cm^3. The filling of the piezometer was executed under vacuum. For the preparation of the mixtures deaerated and doubly distilled water and n-hexane of 99.86 mol% purity were used. The weighting was performed on weight balance machine WLR-200 with an uncertainty of not more than ± 0.5 mg. The measuring error of density ρ makes 0.12-0.15 %.

The detailed features of the apparatus are given in [6].

3. RESULTS AND DISCUSSION

The phase diagram (Fig.1 [7]) of the n-hexane- water system is of type III [8], which means that no critical liquid-gas states occur at concentrations between 0.257 and 0.882 m.f. of water. The low-pressure branch of the critical curve starts in the critical point of pure n-hexane, CPH, and ends in an upper critical endpoint UCEP. The high-pressure branch of the critical line begins in the critical point, CPW, of water, passes through a minimum of temperature and goes towards high pressures [4]. The minimum temperature for our data is 627.7 K, and the pressure is 30.9 MPa. This point is referred to as a double critical point (DCP) and divides the critical line into two parts: the critical line for liquid-gas and the critical line gas-gas [7].

Experimental research of PVT-properties permits us to determine quite precisely the points on phase equilibrium curves of liquid-liquid and liquid-vapor. The typical $P(T)$ diagram for the system n-hexane – water with x = 0.964 m.f. of water along different isochores is presented in Fig.2.

Experimental PVT data for all studied compositions and isochores are presented in Table 1.

Connecting the points of the fractures on the dependencies $P(T)$ the curves of the phase equilibrium liquid-liquid and liquid-vapor were defined.

In Fig.3 are shown the lines of the phase equilibrium liquid-vapor for the system n-hexane – water with all values of X.

All lines of the phase equilibrium liquid-liquid have approximately the forms of parabolas. Therefore we present only one representative curve of phase equilibrium liquid-liquid with X = 0.257 m.f. of water (Fig.4.).

The values of critical parameters T_c, ρ_c, and P_c of phase transitions liquid-liquid and liquid-vapor for the binary n-hexane - water system with the different compositions X are given in Table 2.

For the approximation of the curves of the phase equilibrium liquid-vapor the following expression was used [9,10]

$$\frac{\rho_l - \rho_g}{2\rho_c} = B_0 \left|\tau\right|^{\beta} + B_1 \left|\tau\right|^{\beta+\Delta} , \qquad (1)$$

where $\tau = (T - T_c)/T_c$, ρ_l and ρ_g – are the density of the liquid and gaseous phases, respectively; β, Δ (Δ = 0.50 – fixed value [11]) and B_0, B_1 are the critical indexes and amplitudes. The results of calculations are presented in Table 3 for three compositions (for those, for which one there is the critical point liquid-vapor) of the binary n-hexane – water mixture. As we can see only for one composition (x = 0.257) it is obtained the value of β corresponding to one for the Ising-like system (β = 0.325).

The curves of phase equilibrium liquid-liquid was described by the equation [9,11]

$$\frac{\rho-\rho_c}{\rho_c} = \pm B_0|\tau|^{\beta} \pm B_1|\tau|^{\beta+\Delta} + B_2|\tau|^{1-\alpha} + B_3\tau, \tag{2}$$

where α = 0.11 is the critical exponent that describes the divergence of the specific heat at the critical point; B_2, B_3 are the critical amplitudes. Signs “+” and “-“ are connected with the right and left branch of the curve of the phase equilibrium liquid-liquid, respectively. As demonstrate calculations, for all compositions, the obtained values of β correspond (within the calculating error) to values characteristic for the Ising-like model.

For the description of thermal properties of the n-hexane – water binary system near the critical points liquid-vapor we used the crossover equation of state proposed by Jin et al. [12]. The equation of state incorporates the crossover from singular asymptotic behaviour at the critical point to regular behaviour far away from the critical point and is based on a crossover function introducing the six-term Landau expansion for the Helmholtz free-energy density. Parameters characterising the individual properties of the system were calculated on the basis of literature data [13,14] for pure n-hexane and water and the appropriate PVTx experimental investigations. The dependencies of critical parameters on concentration were approximated by polynomials and spline finctions.

The experimental data on the pressure were compared with the calculated ones. In the region where there is a breaking of the critical line (the curve 3 in the Fig.1) we used critical parameters corresponding to the pseudocritical line for the pressure calculation at x = 0.347, 0.615, and 0.827 m.f. of water.

The equation of state gives us the opportunity to describe the thermodynamic properties (the error is about 3% for pressure description) of the n-hexane – water mixture in the ranges of densities $0.5 \le \rho/\rho_c \le 1.7$ (at $T = T_c$) and temperatures $0.95 \le T/T_c \le 1.3$ (at $\rho = \rho_c$).

ACKNOWLEDGMENTS

This research was supported by the Russian Fundamental Research Foundation (grant N 00-02-17320).

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Table 1. Experimental $PVTx$ properties of binary n-hexane – water system.

T, K	P, MPa	T, K	P, MPa	T, K	P, MPa	T, K	P, MPa
x = 0.166		ρ =334.3 kg/m^3		ρ =202.5 kg/m^3		**x = 0.347**	
ρ = 66.9 kg/m^3		471.75	2.77	482.65	3.57	ρ = 137.5 kg/m^3	
420.01	0.99	472.97	2.82	488.71	3.81	465.45	2.84
422.45	1.02	476.30	2.93	494.21	4.14	466.44	2.92
435.76	1.20	488.45	3.41	502.35	4.62	468.49	2.97
447.65	1.37	496.73	3.68	518.55	5.36	474.67	3.20
460.15	1.62	499.46	3.78	ρ =270.0 kg/m^3		489.55	3.85
469.25	1.85	ρ = 401.2 kg/m^3		492.85	4.93	490.15	3.99
ρ =133.7 kg/m^3		453.90	2.17	493.44	4.95	507.35	5.00
440.51	1.31	458.32	2.30	499.65	5.27	ρ =204.8 kg/m^3	
447.47	1.47	462.95	2.53	ρ =337.5 kg/m^3		487.15	4.46
452.57	1.64	ρ = 468.1 kg/m^3		490.35	5.06	487.86	4.55
459.77	1.83	446.00	1.94	491.92	5.18	489.24	4.74
465.15	1.99	450.25	2.00	494.55	5.28	498.43	5.15
467.29	2.04	ρ = 534.9 kg/m^3		ρ =405.0 kg/m^3		512.19	6.02
472.07	2.22	427.75	1.32	478.75	4.03	519.30	6.28
477.66	2.40	ρ = 601.8 kg/m^3		479.45	4.04	ρ =273.1 kg/m^3	
482.84	2.60	372.75	0.40	488.00	5.18	494.55	5.28
484.04	2.67			493.05	5.60	494.97	5.20
ρ =200.6 kg/m^3		**x = 0.257**		497.81	6.49	497.47	5.39
456.01	1.76	ρ = 135.0 kg/m^3		498.21	6.65	508.95	6.39
458.04	1.83	455.05	2.13	509.36	8.00	521.66	7.26
464.61	2.03	466.57	2.72	510.46	8.30	522.55	7.34
470.15	2.26	477.35	2.97	520.68	9.39	ρ =341.4 kg/m^3	
482.68	2.67	487.14	3.40	533.05	11.18	489.52	4.83
488.45	3.04	509.57	4.41	ρ =435.7 kg/m^3		491.72	5.20
ρ =267.5 kg/m^3		510.14	4.44	446.65	2.15	492.06	5.26
458.90	1.97	510.25	4.51	458.30	2.82	502.98	6.51
470.77	2.37	532.81	5.32	470.65	3.87	513.51	7.89
483.59	2.85	533.05	5.36	ρ =472.5 kg/m^3		525.76	9.76
495.32	3.39	555.00	6.22	437.15	1.88	533.11	10.38
500.45	3.63	572.85	6.93	439.82	1.97	533.15	10.82
ρ =334.3 kg/m^3		573.15	7.02	458.15	3.08	ρ =409.7 kg/m^3	
457.60	2.25	575.03	7.12	ρ =540.0 kg/m^3		477.65	4.03
460.39	2.36	579.75	7.20	432.95	1.67	477.79	4.08

Table 1 (continued)

T, K	P, MPa
x = 0.347	
ρ = 409.7 kg/m^3	
483.76	4.65
488.97	5.33
489.52	5.41
491.77	5.82
492.11	5.89
502.84	7.70
503.13	7.89
516.15	10.24
526.75	12.47
537.12	14.87
548.82	17.12
550.25	17.82
ρ = 477.9 kg/m^3	
462.95	2.98
466.54	3.06
468.99	4.50
469.45	4.62
478.95	6.93
489.75	10.10
509.69	15.80
511.36	15.89
543.97	25.64
550.51	27.33
555.15	29.00
563.13	31.43
565.48	31.93
570.15	33.21
ρ = 546.2 kg/m^3	
428.45	1.40
429.33	1.80
436.67	4.55
443.15	6.89
464.64	15.20
484.06	22.43
507.75	31.60
514.48	34.13
ρ = 546.2 kg/m^3	
525.15	38.11
535.66	42.18
546.49	46.34
556.90	50.13
570.43	55.62
581.25	59.15
x = 0.615	
ρ = 205.1 kg/m^3	
452.90	1.97
459.91	2.38
491.23	4.53
538.48	8.73
562.29	11.39
562.45	11.40
ρ = 273.4 kg/m^3	
464.85	2.52
482.25	3.65
505.87	5.42
524.09	7.41
531.12	8.50
542.15	10.49
550.56	11.73
551.15	11.82
566.05	13.94
570.27	14.51
574.79	15.32
575.15	15.53
ρ = 341.7 kg/m^3	
492.30	5.15
500.48	5.80
511.23	6.84
512.42	6.88
522.65	8.38
534.35	10.21
544.90	11.95
547.28	12.33
ρ = 341.7 kg/m^3	
549.15	12.68
550.65	13.08
578.10	18.81
580.45	19.46
591.65	23.50
ρ = 410.1 kg/m^3	
494.62	5.61
501.80	6.39
502.25	6.43
519.21	9.14
529.72	11.43
530.70	11.54
538.25	13.01
543.77	14.15
591.55	26.61
600.41	28.80
611.65	33.25
ρ = 478.4 kg/m^3	
486.63	4.84
493.90	5.97
509.49	9.20
523.15	12.87
530.71	15.38
531.40	15.60
543.15	19.11
555.35	22.72
564.55	26.43
576.77	30.67
577.35	30.83
586.35	34.20
598.86	38.56
616.80	44.80
625.15	47.75
ρ = 546.8 kg/m^3	
456.35	2.58
460.61	3.01
461.09	3.33
ρ = 546.8 kg/m^3	
466.15	4.46
467.59	4.67
469.37	5.01
473.45	6.21
476.28	6.89
479.05	7.52
482.92	8.77
490.65	11.23
498.15	13.84
506.71	16.65
514.45	19.83
523.15	23.29
548.15	33.69
561.75	38.86
581.05	46.47
582.95	47.51
600.90	55.39
607.25	57.43
ρ = 615.1 kg/m^3	
417.02	1.20
420.55	1.89
437.55	6.67
439.32	7.08
458.90	14.34
461.24	15.02
485.70	25.58
510.31	37.03
536.41	49.88
545.15	54.40
548.65	56.37
553.75	58.83
555.25	59.77
ρ = 649.3 kg/m^3	
395.55	0.88
396.15	1.12
399.65	2.18
402.35	2.85

Table 1 (continued)

T, K	P, MPa
x = 0.615	
ρ = 649.3 kg/m^3	
412.15	6.19
428.65	13.31
450.75	23.44
469.45	32.89
478.65	37.85
480.20	38.69
481.80	39.77
482.25	40.55
496.75	49.88
508.00	55.36
516.60	60.72
524.40	65.34
ρ = 702.4 kg/m^3	
327.05	0.10
329.09	0.75
330.55	1.84
340.99	8.96
342.35	9.67
347.45	12.00
348.45	13.80
352.75	16.57
365.42	24.59
370.25	27.60
378.90	33.42
385.01	37.35
388.35	39.77
393.07	42.88
400.47	47.33
405.05	50.43
412.08	54.51
420.40	60.64
x = 0.827	
ρ = 158.9 kg/m^3	
454.40	2.20
462.74	2.61

T, K	P, MPa
ρ = 158.9 kg/m^3	
474.87	3.25
485.10	3.93
510.14	5.71
534.35	7.43
554.07	9.20
566.95	10.41
ρ = 238.3 kg/m^3	
474.45	3.68
502.36	5.78
525.30	7.87
543.89	9.84
571.65	13.07
580.94	14.89
592.99	17.70
603.85	19.67
ρ = 317.8 kg/m^3	
493.41	5.24
503.67	6.57
513.47	7.37
535.40	9.89
577.02	16.13
595.26	20.30
607.00	22.61
610.15	23.22
ρ = 397.2 kg/m^3	
494.76	5.72
505.42	6.86
527.15	9.58
549.80	13.37
562.33	15.83
583.35	21.24
596.71	25.88
617.25	31.71
ρ = 476.6 kg/m^3	
481.06	4.87
482.47	5.04
491.94	6.12

T, K	P, MPa
ρ = 476.6 kg/m^3	
512.90	9.60
523.00	11.43
532.15	13.28
560.45	20.08
589.05	29.37
601.65	34.89
609.76	37.89
621.65	42.10
ρ = 556.1 kg/m^3	
462.65	3.43
464.61	3.62
478.35	5.60
488.66	7.95
507.74	12.86
522.15	16.68
539.75	22.33
558.25	29.12
572.15	34.04
584.61	39.75
606.99	50.72
614.33	54.20
625.65	60.23
ρ = 635.5 kg/m^3	
447.27	2.63
459.35	5.78
468.76	9.68
482.40	13.60
500.35	21.70
521.97	30.11
534.85	37.47
553.62	46.36
567.99	53.35
582.88	60.28
ρ = 715.0 kg/m^3	
395.65	0.82
406.80	6.83
430.46	17.87

T, K	P, MPa
ρ = 715.0 kg/m^3	
456.90	31.60
472.04	43.21
478.47	48.60
486.20	55.25
492.05	60.15
x = 0.935	
ρ = 176.9 kg/m^3	
433.75	1.39
443.87	1.75
466.35	2.70
470.80	2.85
505.35	4.55
521.45	5.62
542.03	7.53
565.02	9.90
588.95	13.46
613.55	17.44
620.55	18.53
ρ = 265.4 kg/m^3	
454.62	2.19
456.21	2.29
473.27	3.16
487.14	3.67
495.71	4.21
509.07	5.38
533.34	7.61
553.57	9.71
575.87	12.85
591.15	15.38
606.75	18.33
610.25	19.04
624.25	21.79
629.15	22.94
ρ = 353.9 kg/m^3	
464.76	2.58
472.40	3.19

Table 1 (continued)

T, K	P, MPa	T, K	P, MPa	T, K	P, MPa	T, K	P, MPa
x = 0.935		ρ = 530.9 kg/m^3		ρ = 707.8 kg/m^3		ρ = 185.4 kg/m^3	
ρ = 353.9 kg/m^3		517.58	8.07	535.25	20.25	446.22	1.90
480.90	3.85	531.51	9.86	552.64	28.38	457.53	2.20
493.00	5.00	547.88	12.72	571.35	39.22	469.93	2.59
514.01	6.74	567.51	16.57	589.53	51.36	483.99	3.09
545.50	9.84	585.59	20.84	608.48	64.57	495.21	3.61
568.25	12.94	604.67	26.80	ρ = 796.3 kg/m^3		505.93	4.26
591.80	17.36	614.98	30.71	428.41	1.35	530.20	5.89
603.35	19.44	621.15	33.04	429.17	1.70	551.90	7.80
617.35	22.74	ρ = 619.3 kg/m^3		437.94	5.77	586.41	11.76
631.65	25.86	443.51	1.95	447.45	10.53	602.31	14.08
ρ = 442.4 kg/m^3		453.20	2.58	459.46	17.27	607.25	14.72
461.25	2.69	464.10	3.12	467.95	22.37	ρ = 278.1 kg/m^3	
463.12	2.84	470.85	3.55	473.55	26.15	455.15	2.47
474.45	3.62	481.47	4.27	482.35	32.31	466.01	2.89
486.87	4.62	490.37	5.18	495.35	43.50	475.97	3.32
494.00	5.18	502.93	6.80	504.80	52.01	488.35	3.89
495.35	5.36	516.65	9.16	507.12	53.07	511.59	5.26
508.35	6.48	535.86	12.72	518.45	63.54	536.47	7.16
519.15	7.57	562.09	19.58			555.39	9.15
542.85	10.37	578.56	25.16	**x = 0.964**		570.16	10.93
562.85	13.15	584.47	26.75	ρ = 92.7 kg/m^3		592.30	14.86
574.35	15.15	603.93	37.43	418.16	0.83	607.94	16.32
584.86	17.11	615.47	44.22	426.91	1.00	620.65	18.56
585.30	17.25	626.27	50.68	444.19	1.36	628.43	20.07
599.07	20.25	635.42	56.60	459.55	1.74	634.55	21.53
607.15	23.11	646.65	63.55	480.07	2.32	ρ = 370.3 kg/m^3	
614.65	25.38	ρ = 707.8 kg/m^3		497.95	3.08	455.30	2.55
623.15	27.82	433.15	1.43	516.79	4.03	456.25	2.60
630.65	29.61	438.00	1.83	529.30	4.80	466.85	3.18
ρ = 530.9 kg/m^3		449.59	2.31	548.25	6.33	478.46	3.89
454.19	2.40	461.51	3.01	564.94	7.97	503.09	5.35
466.70	3.10	475.26	4.14	575.65	9.12	527.16	7.08
470.35	3.47	482.45	4.75	580.55	9.77	545.04	9.04
476.73	3.95	486.30	5.51	581.45	9.83	565.78	11.46
480.47	4.33	491.69	6.59	ρ = 185.4 kg/m^3		584.06	13.89
489.09	5.08	510.15	11.04	442.15	1.80	600.30	16.70
500.95	6.21	523.76	15.74			614.70	19.58

Table 1 (continued)

T, K	P, MPa
x = 0.964	
ρ = 370.3 kg/m^3	
628.39	22.50
632.15	23.71
ρ = 463.5 kg/m^3	
455.25	2.60
456.25	2.70
466.31	3.18
477.09	4.04
500.63	5.82
525.00	7.89
549.34	10.56
567.50	13.07
583.51	15.68
602.07	19.32
618.40	23.31
621.37	24.14
627.95	25.83
ρ = 556.2 kg/m^3	
455.15	2.58
459.70	2.92
470.25	3.66
485.36	4.76
495.20	5.67
517.95	7.99
530.00	9.37
554.15	12.74
575.73	16.84
597.85	22.29
617.66	28.97
624.25	31.93
626.15	32.70
ρ = 648.9 kg/m^3	
452.05	2.52
454.59	2.64
459.81	2.93
471.53	3.69
483.51	4.66
505.37	7.14
529.49	10.83
544.40	14.15
565.31	19.51
579.21	25.04
598.73	35.08
613.30	43.71
628.15	57.80
ρ = 741.6 kg/m^3	
447.65	2.34
448.48	2.30
456.39	2.70
469.45	3.58
480.30	4.86
492.59	7.10
507.47	11.03
517.20	14.39
535.93	24.72
551.45	35.28
564.60	46.20
575.24	55.22
581.25	60.40
ρ = 834.3 kg/m^3	
425.95	1.39
426.32	1.42
427.30	1.50
432.75	2.08
436.46	3.15
440.34	5.20
445.96	9.87
455.21	17.16
468.62	27.71
480.20	37.30
491.41	46.70
507.26	60.82

Table 2. The values of critical parameters T_c, ρ_c, and P_c of phase transitions liquid-liquid and liquid-vapor for the binary n-hexane - water system with the different compositions x.

x	Liquid-liquid			Liquid-vapor		
	T_c, K	ρ_c, kg/m^3	P_c, MPa	T_c, K	ρ_c, kg/m^3	P_c, MPa
0.166	458.90	267.45	1.97	500.45	267.45	3.63
0.257	491.85	269.98	4.93	494.55	337.05	5.28
0.347	494.55	273.10	5.28	-	-	-
0.615	494.62	410.10	5.61	-	-	-
0.827	494.76	397.20	5.72	-	-	-
0.935	464.76	353.90	2.58	631.65	353.90	25.86
0.964	455.30	370.79	2.55	634.55	278.09	31.53

Table 3. The calculated values of the critical exponent β and critical amplitudes B_0, B_1 for the coexistence curve liquid-vapor of the binary n-hexane – water system with three compositions according equation (1).

x	β	B_0	B_1
0.166	0.901 ± 0.120	13.63 ± 0.78	-
0.257	0.350 ± 0.057	1.564 ± 0.457	-0.908 ± 0.457
0.935	0.553 ± 0.082	4.514 ± 1.229	5.643 ± 2.444

FIGURE CAPTIONS

Fig. 1. The phase diagram for the n-hexane - water system: UCEP, upper critical endpoint; CPH, the critical point of pure n-hexane; CPW, the critical point of pure water; DCP, the double critical point; 1, line of critical point liquid-gas; 2, line of critical point liquid-liquid; 3- pseudocritical line.

Fig.2. The typical $P(T)$ diagram for the system n-hexane – water with x = 0.964 m.f. of water with ρ (kg/m^3) as follows: 1, 92.7; 2, 185.4; 3, 278.1; 4, 370.29; 5, 463.5; 6, 556.2; 7, 648.9; 8, 741.6; 9, 834.3.

Fig.3. The lines of the phase equilibrium liquid-vapor for the system n-hexane – water with x (m.f. of water) as follows: 1, 0.166; 2, 0.257; 3, 0.347; 4, 0.615; 5, 0.827; 6, 0.935; 7, 0.964.

Fig.4. The line of the phase equilibrium liquid-liquid for the system n-hexane – water with x = 0.257 m.f. of water.

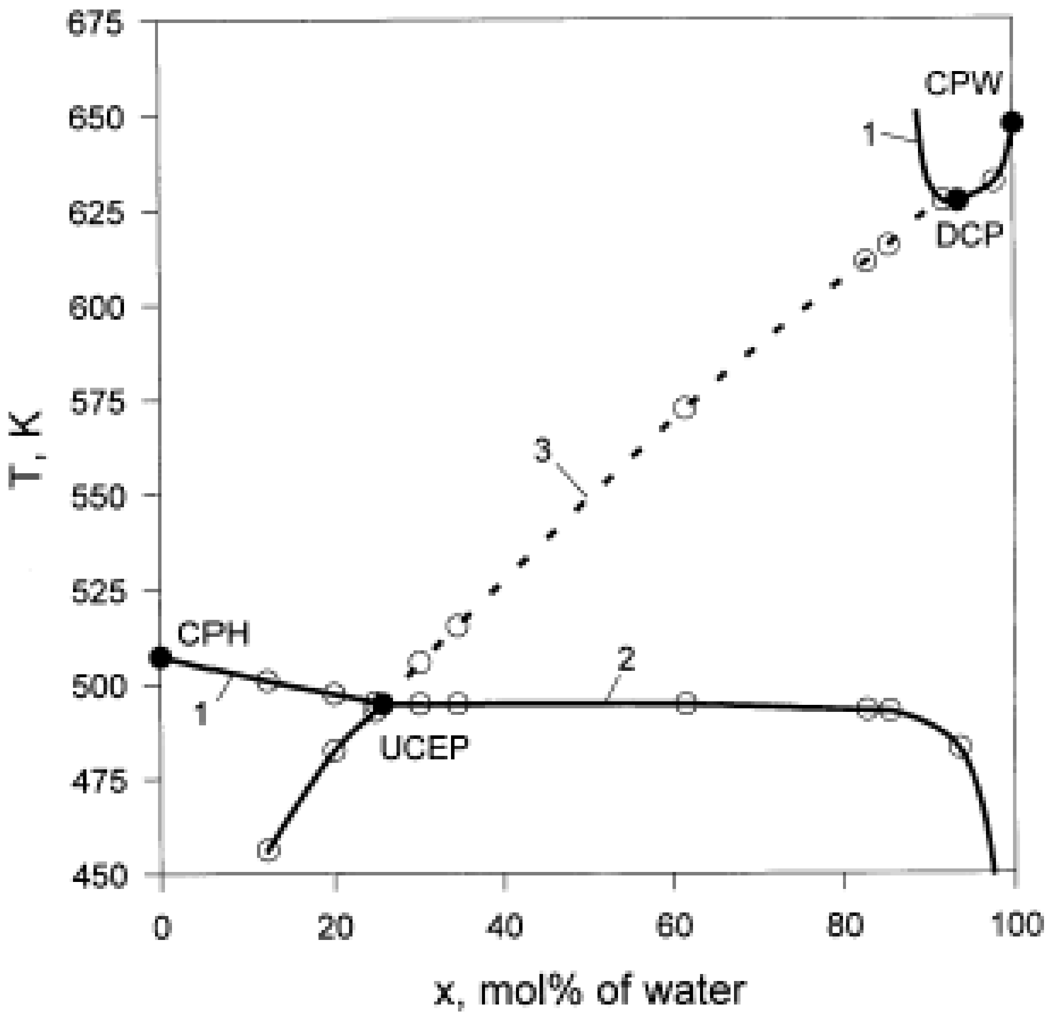
675
650
625
600
575
550
525
500
475
450
T, K
0
20
40
60
80
100
x, mol% of water
CPW
1
DCP
3
CPH
2
1
UCEP

Fig.1.

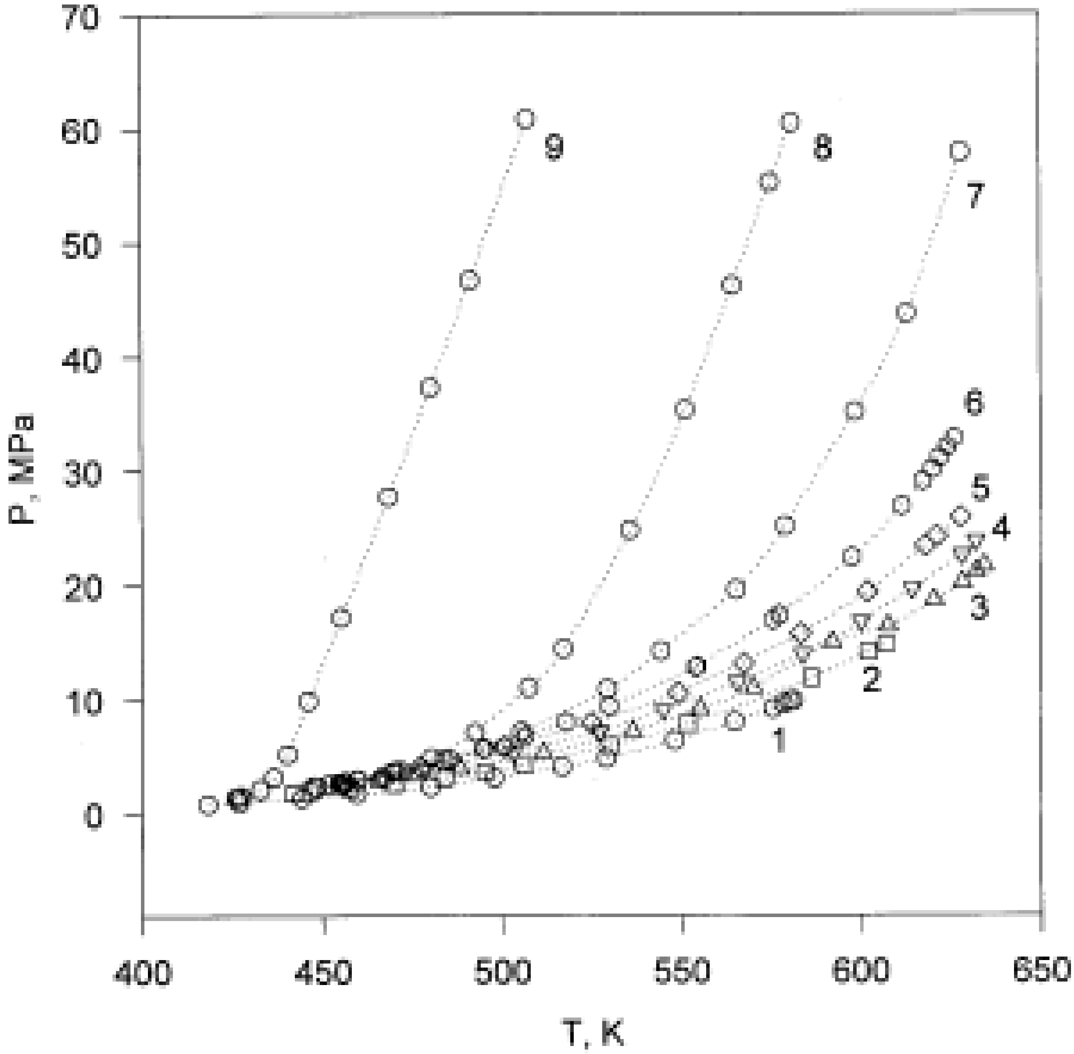

Fig.2.

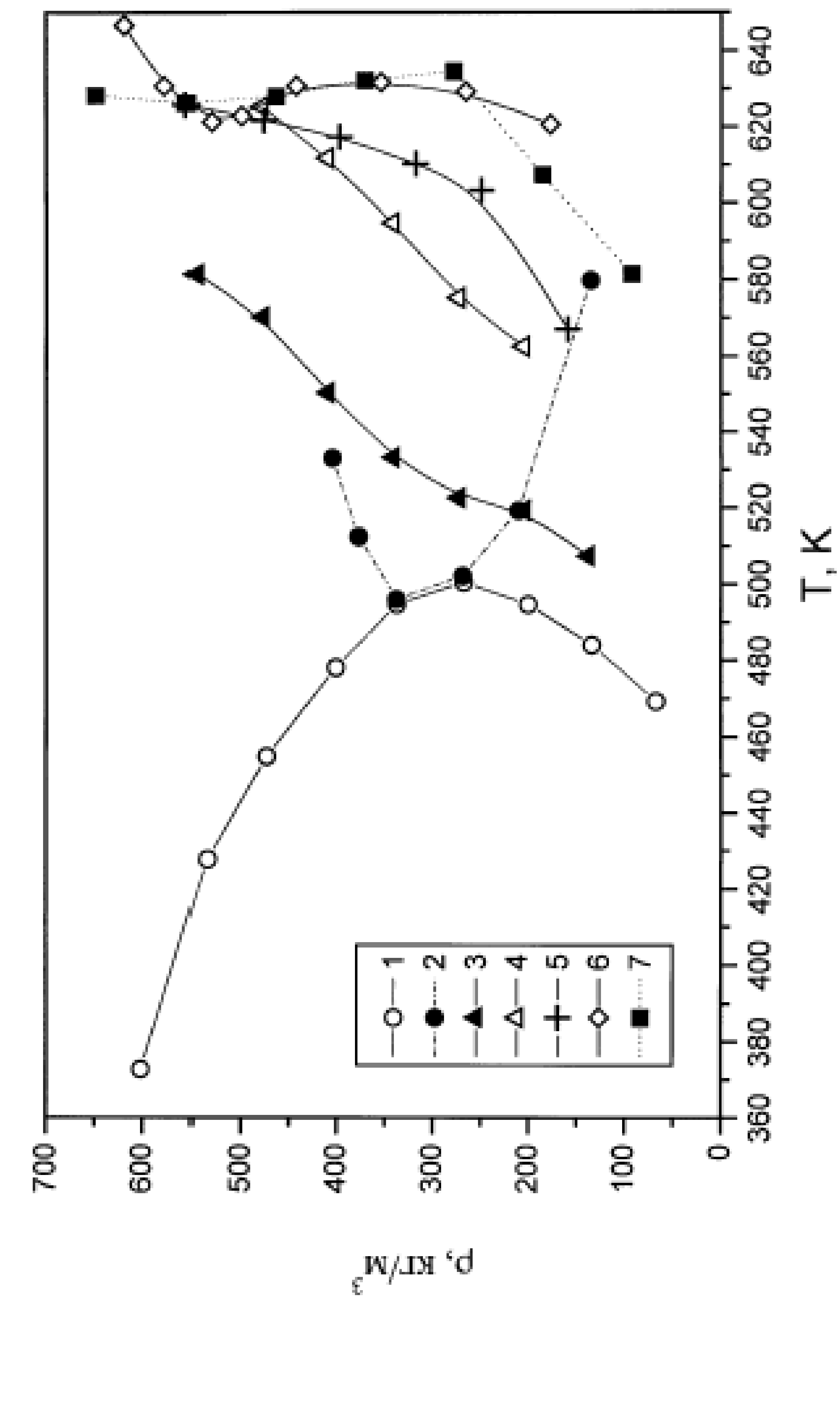
T, K
ρ, кг/м3
1
2
3
4
5
6
7
360
380
400
420
440
460
480
500
520
540
560
580
600
620
640
0
100
200
300
400
500
600
700

Fig.3.

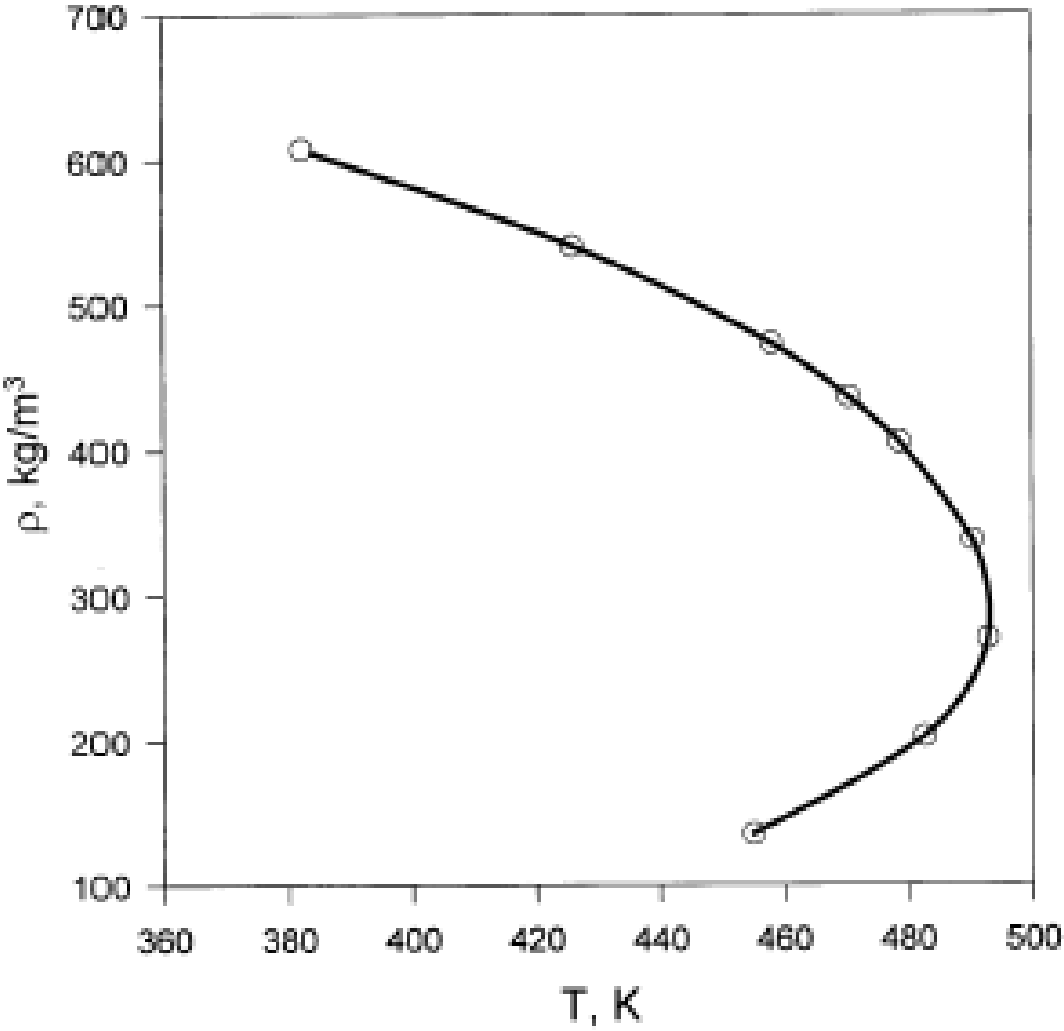
700
600
500
400
300
200
100
ρ, kg/m3
360
380
400
420
440
460
480
500
T, K

Fig.4.